

Application No.: 09/936,184
Amendment Dated: August 7, 2003
Reply to Office Action of: May 7, 2003

This listing of claims will replace all prior versions, and listings, of claims in the application:

LISTING OF CLAIMS

1. (Previously Presented) A a process for the catalytic gas-phase oxidation of propene to acrylic acid, comprising:

passing in which a reaction gas starting mixture 1 which ~~contains propene~~, comprises molecular oxygen and propene in a molar $O_2:C_3H_6$ ratio of ≥ 1 , and at least one inert gas; ~~comprising which comprises~~ at least 20% by volume of molecular nitrogen, ~~and which contains the molecular oxygen and the propene in a molar $O_2:C_3H_6$ ratio of ≥ 1~~ is first passed, in a first reaction stage at elevated temperatures, over a first fixed-bed catalyst, whose active material is at least one multimetal oxide ~~containing~~ comprising molybdenum and/or tungsten and bismuth, tellurium, antimony, tin and/or copper,

thereby obtaining a product gas mixture 1;

wherein said passing of said reaction gas starting mixture 1 proceeds in such a way that the

a propene conversion in a single pass is ≥ 90 mol%, ~~and the~~

an associated selectivity of ~~the~~ an acrolein formation and of ~~the~~ an acrylic acid byproduct formation together is ≥ 90 mol%,

optionally, the a temperature of ~~the~~ said product gas mixture 1 leaving ~~the~~ said first reaction stage is, ~~if required~~, reduced by indirect and/or direct cooling, and, ~~if required~~

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optionally, molecular oxygen and/or inert gas are/is added to the said product gas mixture 1, and

passing said the product gas mixture 1, as reaction gas starting mixture 2 which ~~comprises contains acrolein~~, molecular oxygen and acrolein in a molar $O_2:C_3H_4O$ ratio of ≥ 0.5 , and at least one inert gas, ~~comprising which comprises~~ at least 20% by volume of molecular nitrogen, ~~and which contains the molecular oxygen and the acrolein in a molar $O_2:C_3H_4O$ ratio of ≥ 0.5~~ , is then passed, in a second reaction stage at elevated temperatures, over a second fixed-bed catalyst whose active material is at least one molybdenum- and vanadium-containing multimetal oxide,

thereby obtaining a product gas mixture 2.

wherein said passing of said reaction gas starting mixture 2 proceeds in such away that

an the acrolein conversion in a single pass is ≥ 90 mol%, and the selectivity of the acrylic acid formation balanced over both reaction stages is ≥ 80 mol%, based on propene converted;;

wherein

- a) ~~the a~~ loading of the said first fixed-bed catalyst with the propene contained in reaction gas starting mixture 1 is ≥ 160 l(S.T.P.) of propene/l of catalyst bed h,
- b) ~~the said~~ first fixed-bed catalyst ~~consists of~~ comprises a catalyst bed arranged in two spatially successive ~~reactor~~ reaction zones A, B, ~~the~~

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wherein a temperature of reaction zone A ~~being is~~ from 300 to 390°C and ~~the a~~ a temperature of reaction zone B ~~being is~~ from 305 to 420°C and at the same time at least 5°C above the temperature of reaction zone A,

- c) the reaction gas starting mixture 1 flows first through reaction zone A and then through reaction zone B,
- d) the reaction zone A extends to a propene conversion of from 40 to 80 mol%,
- e) ~~the a~~ loading of ~~the said~~ second fixed-bed catalyst with the acrolein contained in reaction gas starting mixture 2 is ≥ 140 l(S.T.P.) of acrolein/l of catalyst bed \cdot h,
- f) ~~the said~~ second fixed-bed catalyst ~~consists of~~ comprises a catalyst bed arranged in two spatially successive reaction zones C,D,

the

wherein a temperature of reaction zone C ~~being is~~ from 230 to 270°C and ~~the a~~ a temperature of reaction zone D ~~being is~~ from 250 to 300°C and at the same time at least 5°C above the temperature of reaction zone C,

- g) the reaction gas starting mixture 2 flows first through reaction zone C and then through reaction zone D, and
- h) the reaction zone C extends to an acrolein conversion of from 55 to 85 mol%.

2. (Original) A process as claimed in claim 1, wherein the reaction zone A extends to a propene conversion of from 50 to 70 mol%.

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3. (Original) A process as claimed in claim 1, wherein the reaction zone A extends to a propene conversion of from 65 to 75 mol%.

4. (Previously Presented) A process as claimed in claim 1, wherein the reaction zone C extends to an acrolein conversion of from 65 to 80 mol%.

5. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone B is at least 10°C above the temperature of the reaction zone A.

6. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone D is at least 20°C above the temperature of the reaction zone C.

7. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone B is from 305 to 340°C.

8. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone B is from 310 to 330°C.

9. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone C is from 245 to 260°C.

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10. (Previously Presented) A process as claimed in claim 1, wherein the temperature of the reaction zone D is from 265 to 285°C.

11. (Previously Presented) A process as claimed in claim 1, wherein the propene conversion in a single pass in the first reaction stage is ≥ 94 mol%.

12. (Previously Presented) A process as claimed in claim 1, wherein the selectivity of the acrolein formation and of the acrylic acid byproduct formation together in a single pass in the first reaction stage is ≥ 94 mol%.

13. (Previously Presented) A process as claimed in claim 1, wherein the acrolein conversion in a single pass in the second reaction stage is ≥ 94 mol%.

14. (Previously Presented) A process as claimed in claim 1, wherein the selectivity of the acrylic acid formation balanced over both reaction stages is ≥ 85 mol%, based on propene converted.

15. (Previously Presented) A process as claimed in claim 1, wherein the propene loading of the first fixed-bed catalyst is ≥ 165 l(S.T.P.)/l · h.

16. (Previously Presented) A process as claimed in claim 1, wherein the propene

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loading of the first fixed-bed catalyst is $\geq 170 \text{ l(S.T.P.)}/\text{l} \cdot \text{h}$.

17. (Previously Presented) A process as claimed in claim 1, wherein the at least one inert gas contained in the reaction gas starting mixture 1 comprises $\geq 40\%$ by volume of molecular nitrogen.

18. (Previously Presented) A process as claimed in claim 1, wherein the at least one inert gas contained in the reaction gas starting mixture 1 comprises $\geq 60\%$ by volume of molecular nitrogen.

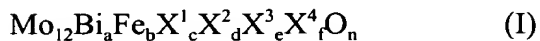
19. (Previously Presented) A process as claimed in claim 1, wherein the at least one inert gas contained in the reaction gas starting mixture 1 comprises steam.

20. (Previously Presented) A process as claimed in claim 1, wherein the at least one inert gas contained in the reaction gas starting mixture 1 comprises CO_2 and/or CO .

21. (Previously Presented) A process as claimed in claim 1, wherein the propene content of the reaction gas starting mixture 1 is from 4 to 15% by volume.

22. (Previously Presented) A process as claimed in claim 1, wherein the active material of the first fixed-bed catalyst is at least one multimetal oxide of the formula I

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where

X^1 is nickel and/or cobalt,

X^2 is thallium, an alkali metal and/or an alkaline earth metal,

X^3 is zinc, phosphorus, arsenic, boron, antimony, tin, cerium, lead and/or tungsten,

X^4 is silicon, aluminum, titanium and/or zirconium,

a is from 0.5 to 5,

b is from 0.01 to 5,

c is from 0 to 10,

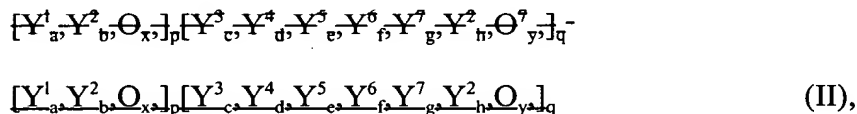
d is from 0 to 2,

e is from 0 to 8,

f is from 0 to 10 and

n is a number which is determined by the valency and frequency of the elements other than oxygen in I.

23. (Currently Amended) A process as claimed in claim 1, wherein the active material of the first fixed-bed catalyst is at least one multimetal oxide of the formula II



where

Y^1 is bismuth, tellurium, antimony, tin and/or copper,

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Y^2 is molybdenum and/or tungsten,

Y^3 is an alkali metal, thallium and/or samarium,

Y^4 is an alkaline earth metal, nickel, cobalt, copper, manganese, zinc, tin, cadmium and/or mercury,

Y^5 is iron, chromium, cerium and/or vanadium,

Y^6 is phosphorus, arsenic, boron and/or antimony,

Y^7 is a rare earth metal, titanium, zirconium, niobium, tantalum, rhenium, ruthenium, rhodium, silver, gold, aluminum, gallium, indium, silicon, germanium, lead, thorium and/or uranium,

a' is from 0.01 to 8,

b' is from 0.1 to 30,

c' is from 0 to 4,

d' is from 0 to 20,

e' is from 0 to 20,

f is from 0 to 6,

g' is from 0 to 15,

h' is from 8 to 16,

x', y' are numbers which are determined by the valency and frequency of the elements other than oxygen in II and

p, q are numbers whose ratio p/q is from 0.1 to 10,

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b₁
containing three-dimensional regions which are delimited from their local environment as a result of their composition differing from their local environment and have the chemical composition Y_a^1, Y_b^2, O_x , and whose maximum diameters are from 1 nm to 100 μm .

24. (Previously Presented) A process as claimed in claim 1, wherein the first fixed-bed catalyst comprises annular and/or spherical catalysts.

25. (Original) A process as claimed in claim 24, wherein the ring geometry is the following:

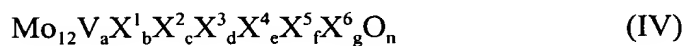
external diameter: from 2 to 10 mm,
length: from 2 to 10 mm,
wall thickness: from 1 to 3 mm.

b₂
26. (Currently Amended) A process as claimed in claim 24, wherein the spherical catalyst is a coated catalyst ~~consisting of~~ comprising a spherical support having a diameter of (from 1 to 8 mm ~~diameter~~) and a coat of active material applied thereon having a thickness of (from 10 to 1000 μm ~~thick~~).

27. (Previously Presented) The process as claimed in claim 1, wherein the first and the second reaction stages are each carried out in a two-zone tube-bundle reactor.

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28. (Previously Presented) A process as claimed in claim 1, wherein the active material of the second fixed-bed catalyst is at least one multimetal oxide of the formula IV



where

X^1 is W, Nb, Ta, Cr and/or Ce,

X^2 is Cu, Ni, Co, Fe, Mn and/or Zn,

X^3 is Sb and/or Bi,

X^4 is one or more alkali metals,

X^5 is one or more alkaline earth metals,

X^6 is Si, Al, Ti and/or Zr,

a is from 1 to 6,

b is from 0.2 to 4,

c is from 0.5 to 18,

d is from 0 to 40,

e is from 0 to 2,

f is from 0 to 4,

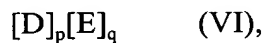
g is from 0 to 40 and

n is a number which is determined by the valency and frequency of the elements other than oxygen in IV.

29. (Previously Presented) A process as claimed in claim 1, wherein the active

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material of the second fixed-bed catalyst is at least one multimetal oxide of the formula VI



where

D is $Mo_{12}V_{a''}Z^1_{b''}Z^2_{c''}Z^3_{d''}Z^4_{e''}Z^5_{f''}Z^6_{g''}O_{x''}$,

E is $Z^7_{12}Cu_hH_{i''}O_{y''}$,

Z^1 is W, Nb, Ta, Cr and/or Ce,

Z^2 is Cu, Ni, Co, Fe, Mn and/or Zn,

Z^3 is Sb and/or Bi,

Z^4 is Li, Na, K, Rb, Cs and/or H,

Z^5 is Mg, Co, Sr and/or Ba,

Z^6 is Si, Al, Ti and/or Zr,

Z^7 is Mo, W, V, Nb and/or Ta,

a'' is from 1 to 8,

b'' is from 0.2 to 5,

c'' is from 0 to 23,

d'' is from 0 to 50,

e'' is from 0 to 2,

f'' is from 0 to 5,

g'' is from 0 to 50,

h'' is from 4 to 30,

i'' is from 0 to 20 and

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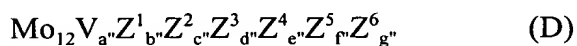
x",y" are numbers which are determined by the valency and frequency of the elements other than oxygen in VI and

p,q are numbers other than zero whose ratio p/q is from 160:1 to 1:1,

which is obtainable by separately preforming a multimetal oxide material (E)



in finely divided form (starting material 1) and then incorporating the preformed solid starting material 1 into an aqueous solution, an aqueous suspension or a finely divided dry blend of sources of the elements Mo, V, Z¹, Z², Z³, Z⁴, Z⁵, Z⁶, which contains the abovementioned elements in the stoichiometry D



(starting material 2), in the desired ratio p:q, drying any resulting aqueous mixture, and calcining the dry precursor material thus obtained, before or after it has been dried, at from 250 to 600°C to give the desired catalyst geometry.

30. (Previously Presented) A process as claimed in claim 1, wherein the second fixed-bed catalyst comprises annular catalysts.

31. (Previously Presented) A process as claimed in claim 1, wherein the second fixed-bed catalyst comprises spherical catalysts.